

Serial No. 09/857,553  
Filed: June 5, 2001

**Remarks/Arguments**

Reconsideration of the application captioned above in view of the above amendments and the below remarks is respectfully requested.

In the Office Action dated March 25, 2003, the Patent Office rejected claims 1, 3-9, and 11-26 under 35 U.S.C. § 102(b) as allegedly being anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as allegedly being obvious over Ota *et al.* (United States Patent No. 5,942,369). For the reasons set forth below, this rejection is traversed.

The Patent Office stated that Ota *et al.* discloses photoresist compositions comprising a mixture of two resins, the first being a novolak resin and the second being an acrylic resin defined in Ota *et al.* in the abstract thereof as well as at column 3, line 9 to column 4, line 44, and in Examples 1 to 12. Applicant disagrees.

The acrylic resins disclosed by Ota *et al.* contain a unit of "a radical-polymerizable compound having an alcoholic hydroxyl group and at least one structural unit selected from the group consisting of a unit of a radical-polymerizable compound having a carboxyl group and a unit of a radical-polymerizable compound having a phenolic hydroxyl group." See column 3, lines 10 to 15. Applicant's claims do not contain such units.

Applicant also submits a Declaration under 37 C.F.R. § 1.132, made by the inventor. Therein, it was found that when 2-hydroxy ethyl methacrylate is not present in (i) a methyl methacrylate/n-butyl methacrylate/acrylic acid system or (ii) a methyl

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methacrylate/acrylic acid system, the film remaining ratio was higher and contrast was improved.

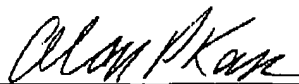
When 2-hydroxy ethyl methacrylate is added into (i) a methyl methacrylate/n-butyl methacrylate/acrylic acid system or (ii) a methyl methacrylate/acrylic acid system, the film remaining ratio became lower and no improvement of contrast was observed.

Given the above, applicant's invention is no longer anticipated by or obvious over Ota *et al.* Withdrawal of the rejections is requested.

Applicant is also filing, under separate facsimile certificates, a three (3) month extension of time, a Notice of Appeal, and an associate power of attorney.

Applicant submits that the concerns of the Patent Office have been addressed. Withdrawal of the rejections and issuance of a Notice of Allowance is respectfully solicited.

Respectfully submitted,



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Attorney's Docket Number: 1999JP311K

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re the application of: Shuichi Takahashi

Serial No.: 09/857,553

Group Art Unit: 1752

Filed: June 5, 2001

Examiner: CHU, John S. Y.

For: RADIATION SENSITIVE COMPOSITION

**Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450**

**DECLARATION UNDER 37 C.F.R. §1.132**

I, Shuichi Takahashi, a Japanese citizen, residing in Kakegawa-shi, Japan, declare as follows:

I received a Bachelor of Engineering degree from Chiba University in 1989 and a Master of Engineering degree in synthetic chemistry from Hokkaido University in 1991. I have been employed by Clariant (Japan) K.K. as a researcher and a Project Leader since 1998. I have been involved in research and development work relating to photoresists and ancillary chemicals since 1998.

I have been named as an inventor on at least 1 United States Patent, 4 US Patent pending and 16 Japanese patent pending applications. I am an applicant in the above-mentioned patent application and an inventor of the invention claimed therein. I am familiar with the references which were used in the rejection thereof.

In order to illustrate the performance of the photoresist compositions of the present invention over compositions which contain a hydroxy acrylate, the following experiments were performed by me or under my direction and control by a colleague who has communicated his findings to me.

**< Synthesis Examples >**

The following acrylic copolymers were prepared in a manner similar to that set forth on pages 36 to 39 of the application captioned above.

**Synthesis Example 1** (PAT-A)

900g of propylene glycol monomethyl ether acetate, 204g of methyl methacrylate, 87g of n-butyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was raised while blowing nitrogen thereinto, and polymerization was conducted at 85°C for 8 hours to obtain an acrylic copolymer of 14,000 in weight average molecular weight and 8mg KOH/g in acid value.

**Synthesis Example 2** (PAT-1)

900g of propylene glycol monomethyl ether acetate, 147g of methyl methacrylate, 60g of n-butyl methacrylate, 84g of 2-hydroxy ethyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was raised while blowing nitrogen thereinto, and polymerization was conducted at 85°C for 8 hours to obtain an acrylic copolymer of 14,700 in weight average molecular weight and 8mg KOH/g in acid value.

**Synthesis Example 3** (PAT-2)

900g of propylene glycol monomethyl ether acetate, 103.5g of methyl methacrylate, 43.5g of n-butyl methacrylate, 144g of 2-hydroxy ethyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was

raised while blowing nitrogen thereinto, and polymerization was conducted at 85°C for 8 hours to obtain an acrylic copolymer of 14,900 in weight average molecular weight and 8mg KOH/g in acid value.

**Synthesis Example 4 (PAT-3)**

900g of propylene glycol monomethyl ether acetate, 291g of methyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was raised while blowing nitrogen thereinto, and polymerization was conducted at 85°C for 8 hours to obtain an acrylic copolymer of 12,900 in weight average molecular weight and 8mg KOH/g in acid value.

**Synthesis Example 5 (PAT-4)**

900g of propylene glycol monomethyl ether acetate, 201g of methyl methacrylate, 90g of 2-hydroxy ethyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was raised while blowing nitrogen thereinto, and polymerization was conducted at 85°C for 8 hours to obtain an acrylic copolymer of 14,500 in weight average molecular weight and 8mg KOH/g in acid value.

**Synthesis Example 6 (PAT-5)**

900g of propylene glycol monomethyl ether acetate, 141g of methyl methacrylate, 150g of 2-hydroxy ethyl methacrylate, 9g of acrylic acid, 6g of azobisdimethylvaleronitrile and 4.5g of azobismethylbutyronitrile were charged into a 2000-ml four-necked flask equipped with a stirrer, a cooling tube, a thermometer and a nitrogen-introducing tube, and stirred. Then, the temperature of the contents was raised while blowing nitrogen

th reinto, and polymerization was conducted at 85°C for 8hours to obtain an acrylic copolymer of 14,600 in weight average molecular weight and 8mg KOH/g in acid value.

The compositions of the Synthesis Examples are summarized in Table 1:

Table 1. Compositions of acrylic copolymers

	monomer ratio (wt%)				Mw
	MMA	n-BMA	2-HEMA	AA	
PAT-A	68	29	0	3	14000
PAT-1	49	20	28	3	14700
PAT-2	34.5	14.5	48	3	14900
PAT-3	97	0	0	3	12900
PAT-4	67	0	30	3	14500
PAT-5	47	0	50	3	14600

MMA : Methyl Methacrylate

n-BMA : n-Butyl Methacrylate

2-HEMA : 2-Hydroxy ethyl methacrylate

AA : Acrylic acid

### <Test Examples>

The following test examples were prepared in a manner similar to that found on pages 39 and 40 of the application captioned above.

#### **Example 1** (PAT-A/polymer=2.25 parts/novolak Mw=8100)

To 100 parts by weight of a novolak resin (weight average molecular weight: 8100) were added 25 parts by weight of an esterification product between 2,3,4,4'-tetrahydroxybenzophenone and 1,2-naphthoquinonediazide-5-sulfonylchloride and 2.25 parts by weight of polymer synthesized in Synthesis Example 1, and the resulting mixture was dissolved in propylene glycol monomethyl ether acetate. After adding thereto 300ppm of a fluorin -containing surfactant of Megafac R-08 (Dai-Nippon Ink & Chemicals, Inc.), the solution was stirred and filtered through a 0.2- $\mu$ m filter to prepare

a radiation sensitive composition. This composition was spin-coated on a 4-inch silicon wafer, and baked on a hot plate at 90°C for 90 seconds to obtain a 1.5- $\mu$ m thick resist layer. This resist layer was exposed by a g and h line stepper (NA=0.1) made by Nikon (FX-604F) through a test pattern with line and space patterns having various line and space widths and a line-to-space width ratio of 1:1, and developed in a 2.38 weight-% aqueous solution of tetramethylammonium hydroxide at 22.5°C for 120 seconds. After completion of the development, thickness of the layer was again measured. Resist layer-remaining ratio was determined according to the following formula:

$$\text{Resist layer-remaining ratio} = [(\text{thickness before development} - \text{thickness after Development}) / \text{thickness before development}] \times 100$$

In addition, an amount of exposure energy enough to resolve a 5- $\mu$ m line-and-space to 1:1 is determined by observation, which was taken as sensitivity. Thus, there were obtained the results shown in Tables 2 and 3 below.

**Example 2** (PAT-A/polymer=5 parts/novolak Mw=8100)

Procedures were conducted in the same manner as in Example 1 except for replacing the amount of added polymer synthesized in Synthesis Example 1 with 5 parts relative to 100 parts by weight of novolak resin.

**Example 3** (PAT-A/polymer=10 parts/novolak Mw=8100)

Procedures were conducted in the same manner as in Example 1 except for replacing the amount of added polymer synthesized in Synthesis Example 1 with 10 parts relative to 100 parts by weight of novolak resin.

**Example 4, 5 and 6** (PAT-1/Table 2)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 2 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 2. In addition, the added amounts of Synthesis Example 2 were as shown in Table 2. Thus, there were obtained the results shown in Table 2.

**Example 7, 8 and 9** (PAT-2/Table 2)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 2 and the

added acrylic copolymer with the polymer synthesized in Synthesis Example 3. In addition, the added amounts of Synthesis Example 3 were as shown in Table 2. Thus, there were obtained the results shown in Table 2.

**Example 10, 11 and 12** (PAT-3/ Table 2)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 2 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 4. In addition, the added amounts of Synthesis Example 4 were as shown in Table 2. Thus, there were obtained the results shown in Table 2.

**Example 13, 14 and 15** (PAT-4/ Table 2)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 2 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 5. In addition, the added amounts of Synthesis Example 5 were as shown in Table 2. Thus, there were obtained the results shown in Table 2.

**Example 16, 17 and 18** (PAT-5/ Table 2)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 2 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 6. In addition, the added amounts of Synthesis Example 6 were as shown in Table 2. Thus, there were obtained the results shown in Table 2.

**Example 19, 20 and 21** (PAT-1/ Table 3)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 3 and the added acrylic copolymer with polymer synthesized in Synthesis Example 2. In addition, the added amounts of Synthesis Example 2 were as shown in Table 3. Thus, there were obtained the results shown in Table 3.

**Example 22, 23 and 24** (PAT-2/ Table 3)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 3 and the



add d acrylic copolymer with th polymer synthesized in Synth sis Example 3. In addition, the added amounts of Synthesis Example 3 were as shown in Table 3. Thus, there were obtained the results shown in Table 3.

**Example 25, 26 and 27** (PAT-3/ Table 3)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 3 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 4. In addition, the added amounts of synthesis Example 4 were as shown in Table 3. Thus, there were obtained the results shown in Table 3.

**Example 28, 29 and 30** (PAT-4/ Table 3)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 3 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 5. In addition, the added amounts of Synthesis Example 5 were as shown in Table 3. Thus, there were obtained the results shown in Table 3.

**Example 31, 32 and 33** (PAT-5/ Table 3)

Procedures were conducted in the same manner as in Example 1 except for replacing the weight average molecular weight of novolak resin as shown in Table 3 and the added acrylic copolymer with the polymer synthesized in Synthesis Example 6. In addition, the added amounts of Synthesis Example 6 were as shown in Table 3. Thus, there were obtained the results shown in Table 3.

In order to compare the contrast of each condition, sensitivity is made the same by changing molecular weight (Mw) of novolak resin. Sensitivity and film remaining ratio were measured by using the novolak resin with the same Mw

## 1. Comparison of contrast by film remaining ratio by adjusting sensitivity with Mw of novolak resin

Table 2. Test Results of Photosensitivity and Film Remaining Ratio

	Loading of Acrylic Resin=2.25				Loading of Acrylic Resin=5.0				Loading of Acrylic Resin=10.0			
	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)
PAT-A	Ex.1	8100	10.6	91.1	Ex.2	6200	12.4	95.3	Ex.3	4200	12.4	92.3
PAT-1	Ex.4	8550	10.6	90.3	Ex.5	7060	12.4	94.0	Ex.6	5100	12.4	91.0
PAT-2	Ex.7	9360	10.6	89.5	Ex.8	8120	12.4	93.2	Ex.9	6180	12.4	90.6
PAT-3	Ex.10	8460	10.6	90.6	Ex.11	7000	14.8	95.2	Ex.12	5000	14.2	93.1
PAT-4	Ex.13	9180	10.6	89.9	Ex.14	8070	14.8	93.8	Ex.15	6160	14.2	91.0
PAT-5	Ex.16	9900	10.6	90.3	Ex.17	9100	14.8	94.0	Ex.18	7260	14.2	91.3

## 2. Comparison of sensitivity and film remaining ratio at the constant Mw of novolak resin

Table 3. Test Results of Photosensitivity and Film Remaining Ratio

	Loading of Acrylic Resin=2.25				Loading of Acrylic Resin=5.0				Loading of Acrylic Resin=10.0			
	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)	Example No.	Mw of Resin	Sensitivity (mJ/cm <sup>2</sup> )	Film Remain. Ratio(%)
PAT-A	Ex.1	8100	10.6	91.1	Ex.2	6200	12.4	95.3	Ex.3	4200	12.4	92.3
PAT-1	Ex.19	8100	below 8	85.4	Ex.20	6200	11.2	92.7	Ex.21	4200	10.6	87.3
PAT-2	Ex.22	8100	below 8	69.5	Ex.23	6200	10.0	84.2	Ex.24	4200	below 8	72.7
PAT-3	Ex.25	9000	13.0	94.1	Ex.26	7000	14.8	95.2	Ex.27	5000	14.2	93.1
PAT-4	Ex.28	9000	10.6	86.5	Ex.29	7000	11.8	88.0	Ex.30	5000	10.0	85.6
PAT-5	Ex.31	9000	below 8	77.1	Ex.32	7000	below 8	81.2	Ex.33	5000	below 8	75.3

When 2-hydroxy ethyl methacrylate is not present in a methyl methacrylate/n-butyl methacrylate/acrylic acid system (PAT-A), the film remaining ratio was higher and contrast was improved.

When 2-hydroxy ethyl methacrylate is added into a methyl methacrylate/n-butyl methacrylate/acrylic acid system (PAT-1; PAT-2), the film remaining ratio became lower and no improvement of contrast was observed.

When 2-hydroxy ethyl methacrylate is not present in a methyl methacrylate/acrylic acid system (PAT-3), the film remaining ratio was higher and contrast was improved.

When 2-hydroxy ethyl methacrylate is added into a methyl methacrylate/acrylic acid system (PAT-4; PAT-5), the film remaining ratio became lower and no improvement of contrast was observed.

The above results were unexpected.

I further declare that all statements herein made of my own knowledge are true and all statements herein made on information and belief are believed to be true. I understand that willful false statements and the like are punishable by fine or imprisonment or both (18 U.S.C. 1001) and may jeopardize the validity of the application or any patent issuing thereon.

June 10, 2003

(date)

S. Takahashi

Shuichi Takahashi